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EARLY STAGES OF *p*-PHENYLENEDIAMINE ANTIOZONANTS REACTION WITH OZONE: RADICAL CATION AND NITROXYL RADICAL FORMATION

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Abstract

Physical-chemical considerations suggest that the diene rubber olefinic double bonds protection by *N,N'*-substituted *p*-phenylenediamines (PPDs) antiozonants (AOZs) is due to their very low ionization potential in comparison to the ionization potential of the olefinic double bonds. Thus, the ozone scavenging action exerted by PPDs AOZs is due to one electron transfer from the PPDs to the ozone molecule with the formation of the PPDs radical cation. Furthermore, Hammett-type plot of the reaction rate with ozone of a series of aromatic amines including also *N,N'*-substituted PPDs suggests that the result of ozone oxidation is the formation of PPDs radical cations. In the present study, a series of common *N,N'*-substituted PPDs used as AOZs in rubber compounds were oxidized with ozone in trichloromethane and/or *o*-dichlorobenzene solutions and studied with Electron Spin Resonance (ESR). Experimental evidences about the radical cation formation and the subsequent almost quantitative PPDs nitroxyl radical formation as a result of the reaction of PPDs radical cation with ozone radical anion were collected and support experimentally the physical-chemical premises. Upon addition of trichloroacetic acid to the PPDs nitroxyl radical solutions, the immediate decomposition of the nitroxyl radical and the formation of stabilized PPDs radical cation was observed.

Key words: Antiozonants; *N,N'*-substituted *p*-phenylenediamine; ozone; ESR; radical cation; nitroxyl radical.

1. Introduction

The ozone concentration in the troposphere and at ground level was measured at about 10 ppb in the XIX century in western Europe [1]. Since then, a steady growth in the ozone concentration in the troposphere was observed and at the beginning of the XXI century the background tropospheric ozone concentration reached 35-45 ppb in the Northern Hemisphere with annual maxima above 110

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